

Spin gap of the three-leg $S = 3/2$ Heisenberg tube

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The ground-state properties of the three-leg $S = 3/2$ Heisenberg tube are studied using the density-matrix renormalization group method. We find that the spin-excitation gap associated with a spontaneous dimerization opens for the whole coupling regime, as seen in the three-leg $S = 1/2$ Heisenberg tube. However, in contrast to the case of $S = 1/2$ tube, the gap increases very slowly with increasing the rung coupling and its size is only a few % or less of the leg exchange interaction in the weak- and intermediate-coupling regimes. We thus argue that, unless the rung coupling is substantially larger than the leg coupling, the gap may be quite hard to be observed experimentally. We also calculate the quantized Berry phase to show that there exist three kinds of valence-bond-solid states depending on the ratio of leg and rung couplings.

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For a long time, the exotic phenomena emerged from geometrical frustration have been fascinating but challenging subjects of research in condensed matter physics.¹ The peculiar dilemma of frustrated systems generally comes from a highly-degenerate ground state in the classical sense. Here, we know that to resolve it comes essentially back to how the degeneracy is removed or how the frustration is minimized by taking the quantum fluctuations into account. The simplest example for the spin frustration may be the 120° structure of antiferromagnetic triangle. In the context of triangular-lattice $S = 1/2$ antiferromagnet a spin-liquid state was proposed by Anderson.² As a related issue, odd-leg spin tube systems such as $\text{Na}_2\text{V}_3\text{O}_7$ (Ref. 3) and $[(\text{CuCl}_2\text{tachH})_3\text{Cl}]\text{Cl}_2$ (Ref. 4) have attracted much attention for the last few years. One could say that odd-leg spin ladders belong to the same universality class as single chain does; thus, the ground state is comprehended as a gapless spin liquid or a Tomonaga-Luttinger (TL) liquid.⁵ However, if the periodic boundary conditions (PBC) are applied in the rung direction, i.e., a tube is shaped, the spin states are dramatically changed due to spin frustration in the polygonal ring with odd number of rungs.

Quite recently, the hexagonal compound CsCrF_4 (Ref. 6), which is an ideal three-leg spin tube system formed by Cr^{3+} ions, has been reexamined experimentally from the point of view of spin frustration.⁷ Since the magnetic moment comes from the e_g^2 state of Cr^{3+} ion, the magnitude of spin on each site is $S = 3/2$. By performing magnetic susceptibility, heat capacity $C(T)$, and electron spin resonance measurements, it was reported that no magnetic long-range order is observed down to $T = 1.3\text{K}$. In particular, a gapless spin-liquid state (or a TL liquid state) is indicated from the finite T -linear component of $C(T)$; this result raises a more absorbing question because a gapped ground state is expected in odd-leg spin-half-integer spin tube system.⁸ The need for an investigation of odd-leg spin tube system with $S = 3/2$

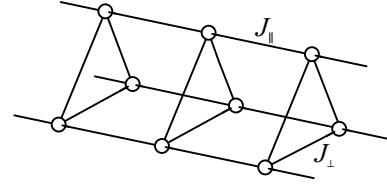


FIG. 1. Lattice structure of three-leg Heisenberg tube.

is therefore obvious in order to figure out this puzzle. In this paper, we thus consider three-leg $S = 3/2$ Heisenberg tube. The Hamiltonian is given as

$$H = J_{\parallel} \sum_{\alpha=1}^3 \sum_i \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha,i+1} + J_{\perp} \sum_i \sum_{\alpha(\neq\alpha')} \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha',i}, \quad (1)$$

where $\vec{S}_{\alpha,i}$ is a spin- $3/2$ operator at leg $\alpha (= 1, 2, 3)$ and rung i . J_{\parallel} and J_{\perp} are the nearest-neighbor exchange interactions in the leg and rung directions, respectively (see Fig.1). We take $J_{\parallel} = 1$ as the unit of energy hereafter. In order to investigate the ground-state properties of the system (1), the density-matrix renormalization group (DMRG) technique⁹ is employed. As necessary, the PBC or the open-end boundary conditions (OBC) are chosen in the leg direction. Using the OBC (PBC), we study the tubes with several kinds of length $L = 12$ to 48 ($L = 8$ to 24) keeping $m = 1200$ to 2600 ($m = 1600$ to 3200) density-matrix eigenstates in the renormalization procedure; in this way, the typical truncation error, i.e., the discarded weight, is $2 \times 10^{-6} - 1 \times 10^{-5}$ ($3 \times 10^{-5} - 7 \times 10^{-5}$). We note that the system length must be even and is better to be kept in $L = 4l$ or $4l + 2$ with l =integer for systematic extrapolation of calculated quantities into the thermodynamic limit. Moreover, an extrapolation to $m \rightarrow \infty$ for each calculation is necessary because the DMRG trial wave function slowly approaches the exact one with increasing m due to the large degrees

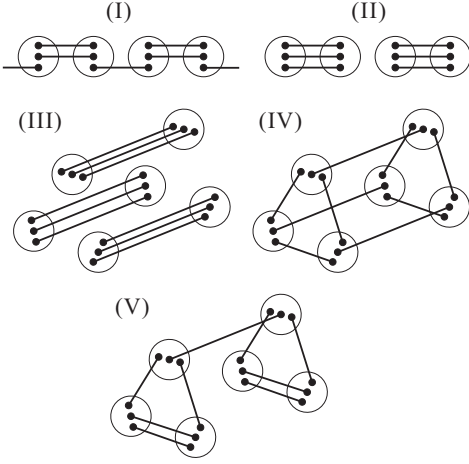


FIG. 2. Schematic pictures of the VBS configurations. Each small solid circle and connection with line denote a spin- $\frac{1}{2}$ variable and a singlet pair, respectively. The large open circle represents a spin- $\frac{3}{2}$ operation which symmetrizes three spin- $\frac{1}{2}$ variables. The configurations (I) and (II) are VBS states in small and large bond-alternation regimes of the 1D $S = \frac{3}{2}$ Heisenberg model, respectively. The configurations (III)-(V) are possible candidates of VBS states in the three-leg $S = \frac{3}{2}$ Heisenberg tube (details are given in the text and Fig. 5).

of freedom $\sim 4^{3L}$ and strong spin frustration in our system. All the calculated quantities in this paper are extrapolated to the limit $m \rightarrow \infty$; thus, for example, the maximum error in the ground-state energy is estimated to be less than 1×10^{-3} .

The first thing we think of when considering the system (1) might be the topological similarity to the other odd-leg half-integer-spin Heisenberg tubes. The simplest case, i.e., three-leg $S = \frac{1}{2}$ Heisenberg tube, has been well-studied, and the ground state is known to be gapped where the system is spontaneously dimerized in the leg direction to relax the intra-rung spin frustration.^{8,10-12} This can be naturally understood by analogy with the gap-opening mechanism in the one-dimensional (1D) $S = \frac{1}{2}$ spin-Peierls Heisenberg model.¹³ Hence, in the case of $S = \frac{3}{2}$ as well, it would be best to start with bond-alternated single chain problem, namely, the 1D $S = \frac{3}{2}$ spin-Peierls Heisenberg model. The Hamiltonian is written as $H = \sum_{i=1}^{L-1} [1 - (-1)^i \delta] \vec{S}_i \cdot \vec{S}_{i+1}$ where \vec{S}_i is a spin- $\frac{3}{2}$ operator at site i and $\delta (> 0)$ is the strength of bond alternation. The low-energy physics of this system has been fundamentally elucidated.^{15,16} Across the critical point $\delta \approx 0.42$, two kinds of valence-bond-solid (VBS) phases appear in the ground state; for the larger alternation ($\delta > 0.42$), the VBS state is essentially written as a direct product of ‘simple’ spin-Peierls singlet bonds [Fig. 2(II)], whereas for the smaller alternation ($\delta < 0.42$), it is expressed as a combined state of the spin-Peierls singlet and $S = 1$ Haldane-like-gapped configurations [Fig. 2(I)]. And, the ground state is always gapped except at the critical point. Now therefore, getting back to our system (1), if the spontaneous dimeriza-

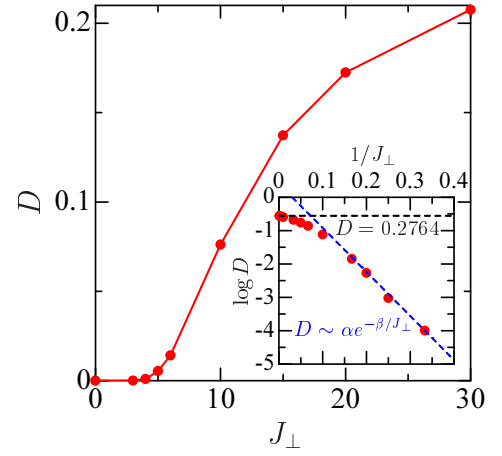


FIG. 3. Dimerization order parameter D as a function of J_{\perp} . Inset: Semilog plot of D vs. $1/J_{\perp}$. D saturates to 0.2764 in the large J_{\perp} limit. The data for small J_{\perp} is fitted by a function $D = \alpha \exp(-\beta/J_{\perp})$ with $\alpha = 0.25$ and $\beta = 13.2$.

tion occurs as in the three-leg $S = \frac{1}{2}$ Heisenberg tube, it is likely that a gapped ground state is also obtained here.

Then, we will simply evaluate a dimerization order parameter to check the presence or absence of long-ranged spin-Peierls ordering in our system. Since a spin-Peierls state is characterized as an alternating formation of spin-singlet pairs in the leg direction, we focus on the nearest-neighbor spin-spin correlations,

$$S(i) = -\langle \vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha,i+1} \rangle, \quad (2)$$

where $\langle \dots \rangle$ denotes an expectation value in the ground state. Note that this quantity is independent of α . With applying the OBC, the translational symmetry is broken due to the Friedel oscillation and the spin-Peierls state is directly observable as a ground state. In general, the Friedel oscillation in the center of the system decays as a function of the system length. If the amplitude at the center of the system

$$D(L) = |S(L/2) - S(L/2 + 1)| \quad (3)$$

persists for arbitrarily long system length, it corresponds to a long-ranged dimerization order which indicates the spin-Peierls ground state. The order parameter is thus defined as an extrapolated value into the thermodynamic limit,

$$D = \lim_{L \rightarrow \infty} D(L). \quad (4)$$

In Fig. 3, the extrapolated values D are plotted as a function of J_{\perp} . We see that it increases gradually at $J_{\perp} \lesssim 5$, almost linearly at $5 \lesssim J_{\perp} \lesssim 15$, and then go into saturation at $D = 0.2764$ in the large J_{\perp} limit. These different behaviors could be interpreted in terms of different VBS state for each the J_{\perp} regime, as in the $S = \frac{1}{2}$ Heisenberg tube.¹¹ This point will be clarified below by examining the Berry phase. A remaining question

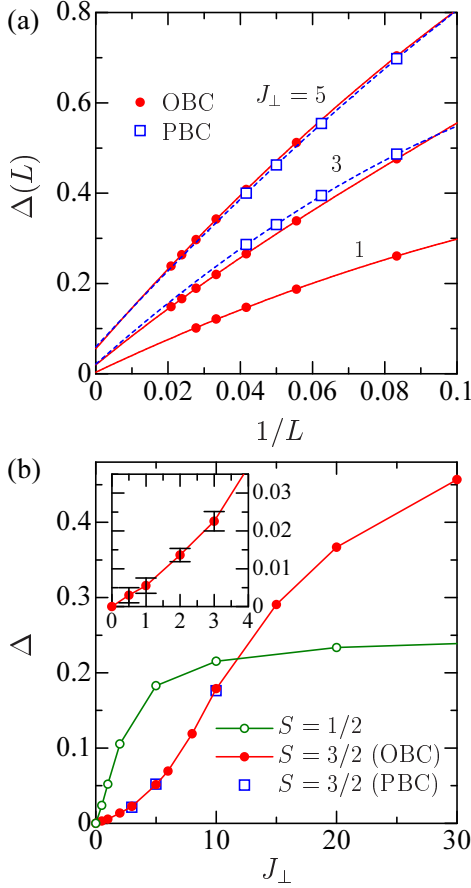


FIG. 4. (a) Finite-size scaling of $\Delta(L)$ as a function of $1/L$. The lines are the polynomial fittings. (b) Extrapolated values of $\Delta(L)$ to the thermodynamic limit $1/L \rightarrow 0$. Inset: extended figure for $0 \leq J_{\perp} \leq 4$. The error bars give differences between the second-order and cubic polynomial fittings for $\Delta(L)$.

would be whether the order parameter remains finite for small J_{\perp} regime (we have not successfully obtained D for $J_{\perp} < 3$ due to its smallness). We find that the behavior seems just like the Berezinskii-Kosterlitz-Thouless type transition, $D = 0.25 \exp(-13.2/J_{\perp})$, from the single logarithmic plot (see the inset of Fig. 3); it may imply that the order parameter is exponentially small but finite at $0 < J_{\perp} < 3$. We conclude that in the wide range of J_{\perp} the dimerization order occurs and the spin-excitation gap is expected to be finite there.

Let us then estimate the spin-excitation gap. Of particular interest is the evolution of the gap onto the ratio between leg and rung couplings. The gap is defined as

$$\Delta = E_1(L) - E_0(L), \quad \Delta = \lim_{L \rightarrow \infty} \Delta(L), \quad (5)$$

where $E_0(L)$ and $E_1(L)$ are energies of the ground state ($S = 0$) and first triplet excited state ($S = 1$) for the system with length L , respectively. In Fig. 4(a), we plot the system-size dependence of the spin-excitation gap calculated with the OBC in full circles. We see

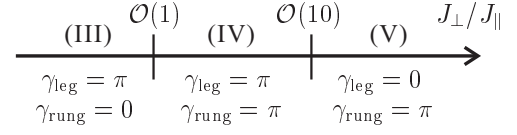


FIG. 5. Schematic phase diagram of the three-leg $S = \frac{3}{2}$ Heisenberg tube, classified by the Berry phases on the leg bond (γ_{leg}) and rung bond (γ_{rung}). The Roman numbers correspond to the VBS states shown in Fig. 2.

that the values of $\Delta(L)$ can be smoothly extrapolated to the thermodynamic limit $1/L \rightarrow \infty$. The extrapolated values Δ , using a cubic polynomial extrapolation for $\Delta(L)$, are shown in Fig. 4(b) as a function of J_{\perp} . As expected, J_{\perp} -dependence of Δ looks rather similar to that of D ; namely, it increases slowly at $J_{\perp} \lesssim 5$, rapidly at $5 \lesssim J_{\perp} \lesssim 15$, and then saturates to $\Delta = 0.6661$ in the strong-coupling limit $J_{\perp} = \infty$. This is because the spin-excitation gap is essentially equivalent to a binding energy of most weakly bounded spin-singlet pair in the system and it is approximately scaled with the dimerization strength.

Here, it is very instructive to compare the gap with that of the $S = \frac{1}{2}$ tube, which is also shown in Fig. 4(b). Two remarks are made by the comparison: (i) Although it may be rather natural, the gap in the strong-coupling limit seems to be scaled with the magnitude of spin, $\Delta(J_{\perp} \rightarrow \infty) \propto S$, (ii) the gap for $S = \frac{3}{2}$ increases much more slowly with increasing J_{\perp} in the weak-coupling regime. As a result, only a few % of the leg exchange interaction remains even at $J_{\perp}/J_{\parallel} = 5$. Hence, we argue that, unless the ratio J_{\perp}/J_{\parallel} is sizably large, it may be difficult to detect the gap experimentally. For CsCrF_4 , the leg exchange interaction is estimated to be a few 10 to 100 K by comparing the experimental peak position of magnetic susceptibility to numerical analysis for the 1D $S = \frac{3}{2}$ Heisenberg chain,¹⁷ and the gap is only a few K at the outside even for $J_{\perp}/J_{\parallel} = 5$.

As described above, we obtain the gapped ground state for the whole J_{\perp} region with applying the OBC. It would mean that our system never contain the Haldane-type VBS state [Fig. 2 (I)] in the three chains. This is because the gap cannot open due to free edge spins created with the OBC if the state (I) is included. But to be sure, we shall confirm it by estimating the gap with the PBC. The obtained results are shown with open squares in Fig. 4. We see that the extrapolated values are in good agreement to those with the OBC and it is confirmed that the state (I) does not exist at any VBS state in our system. Now, it is a fair question then to ask which kind of VBS state is formed and how it changes with varying J_{\perp} .

Finally, we investigate the quantized Berry phase for determining topological configuration of VBS ground state. The Berry phase is defined by

$$\gamma = -i \int_0^{2\pi} A(\phi) d\phi, \quad (6)$$

where $A(\phi)$ is the Abelian Berry connection, $A(\phi) = \langle \psi_\phi | \partial_\phi \psi_\phi \rangle$ with the ground state $|\psi_\phi\rangle$.¹⁸ The Berry phase is quantized as 0 or $\pi \pmod{2\pi}$ if the system has spin gap during the adiabatic continuation and time reversal symmetry; and “undefined” if a gapless excitation exists. We introduce a local perturbation by a local twist of the nearest-neighbor connection, $\vec{S}_{\alpha,i} \cdot \vec{S}_{\alpha',j} \rightarrow \frac{1}{2}(e^{-i\phi} S_{\alpha,i}^+ S_{\alpha',j}^- + e^{i\phi} S_{\alpha,i}^- S_{\alpha',j}^+) + S_{\alpha,i}^z S_{\alpha',j}^z$. In this paper, a couple of clusters with $L = 2$ and 4 are analyzed for this quantity. A dimerized pair of triangles, including six spins, from the clusters are picked up (it is the cluster itself for $L = 2$), and the Berry phases of the leg bond (γ_{leg}) for $\alpha = \alpha', j = i + 1$ and of the rung bond (γ_{rung}) for $\alpha \neq \alpha', j = i$ are evaluated. We call the spin-singlet pair on the leg (rung) bond “on-leg (on-rung) pair”.

The Berry phases are obtained as follows: $(\gamma_{\text{leg}}, \gamma_{\text{rung}}) = (\pi, 0)$ at $0 < J_\perp < 1$ ($0 < J_\perp < 0.5$), $(\gamma_{\text{leg}}, \gamma_{\text{rung}}) = (\pi, \pi)$ at $1 < J_\perp < 15.3$ ($1 < J_\perp < 18$), and $(\gamma_{\text{leg}}, \gamma_{\text{rung}}) = (0, \pi)$ at $J_\perp > 15.3$ ($J_\perp > 18$) for $L = 2$ ($L = 4$) cluster. Accordingly, we find three different phases depending on J_\perp/J_\parallel , as shown in Fig. 5. The term “phase transition” describes a recombination of VBS bonds. In the large-coupling regime $J_\perp/J_\parallel > \mathcal{O}(10)$, we can easily imagine that the on-rung pair is more stable than the on-leg pair and as many pairs as possible prefer to be formed on the rung bond [Fig. 2(V)]. The spin gap is therefore scaled with the binding energy of on-leg pair, i.e., $\Delta \propto J_\parallel$, which is consistent with the saturating behavior of Δ for $J_\perp \gg J_\parallel$. On the other hand, in the weak-coupling regime $J_\perp/J_\parallel < \mathcal{O}(1)$, all spin-singlet pairs are formed on the leg bond [Fig. 2(III)] because the binding energy of the on-leg pair is much larger than that of the on-rung pair. And, in the intermediate regime $\mathcal{O}(1) < J_\perp/J_\parallel < \mathcal{O}(10)$, the spin-singlet pairs seem to

be distributed *in a balanced manner* [Fig. 2(IV)]. Here, we notice an interesting relation to the phase transition in the $S = \frac{1}{2}$ tube.¹⁹ If we ignore a spin-singlet pair on each rung in the phases (IV) and (V) of our system, the remaining degrees of freedom are equivalent to those of the $S = \frac{1}{2}$ tube. As it turns out, the recombination of the remaining VBS bonds between (IV) and (V) can be essentially equivalent to the phase transition in the $S = \frac{1}{2}$ tube. Then the (ignored) extra degrees of freedom yields the additional phase (III) in our $S = \frac{3}{2}$ system.

In conclusion, we study the ground-state properties of the three-leg $S = 3/2$ Heisenberg tube with the DMRG method. It is confirmed that a spontaneous dimerization occurs and the spin-excitation gap opens for the whole coupling region. This may be a common feature of odd-leg half-integer-spin Heisenberg tube systems. We find that the gap for $S = \frac{3}{2}$ increases very slowly with increasing J_\perp and it remains very small compared with J_\parallel in the weak- to intermediate-coupling regions. For CsCrF_4 , the gap is estimated to be only a few K or less at normal pressures and, for example, additional condition such as applying pressure might be required to enlarge the ratio J_\perp/J_\parallel in order to detect the gapped state. Moreover, by calculating the quantized Berry phase, it is shown that two phase transitions as recombination of VBS bonds occur with varying the ratio J_\perp/J_\parallel although further work is desirable for quantitative evaluation of the critical points of the phase transitions.

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